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EFFECTS OF TEMPERATURE AND PRESSURE ON THE EXPLOSIBILITY OF METHANE-AIR MIXTURES

BY

G. A. BURRELL AND I. W. ROBERTSON

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EFFECTS OF TEMPERATURE AND PRESSURE ON THE EXPLOSIBILITY OF METHANE-AIR MIXTURES.

By G. A. Burrell and I. W. Robertson.

INTRODUCTION.

The results of experiments to determine the effect of temperature and pressure on the low limit of complete propagation of flame in mixtures of methane and air are described in this paper. Temperatures up to 500° C. and pressures of 5 atmospheres above atmospheric pressure and 250 mm. below atmospheric pressure were employed.

Experiments to determine the limits of inflammability of methane-air mixtures at ordinary temperatures and pressures and to determine the effects of adding inert gases to such mixtures have been described in other publications[a] of the Bureau of Mines.

METHOD OF CONDUCTING TESTS.

The apparatus used is shown in figure 1. The explosion pipette, *a*, had a capacity of 100 c. c. Platinum wires were fused into the upper part. A spark from an induction coil, driven by four dry cells, was used to ignite the gas mixtures. An electrically heated oven, *d*, surrounded the explosion pipette. The temperatures were measured by means of a platinum-rhodium thermocouple. Transparent mica plates were used to close the upper end of the oven in order to observe the effects of sparking the mixtures. A reserve pipette, *b*, of 200 c. c. capacity was fastened to the explosion pipette by means of pressure rubber tubing, with mercury as the confining fluid.

The apparatus was made ready for use by raising the leveling bottle *c*, thereby completely filling the pipettes *a* and *b* with mercury, and then lowering it until all of the mercury had fallen from *a*, leaving a vacuum therein. The stopcock between the two pipettes was then closed and the required gas mixture drawn into *b* through the free

[a] Burrell, G. A., and Seibert, F. M., The inflammable gases in mine air: Tech. Paper 39, 1913, p. 22; Burrell, G. A., and Oberfell, G. G., The limits of inflammability of mixtures of methane and air: Tech. Paper 119, 1915, 30 pp.; Clement, J. K., The influence of inert gases on inflammable gaseous mixtures: Tech. Paper 43, 1913, 24 pp.

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end of the stopcock. After the oven was heated to the required temperature, the gas mixture was admitted into *a* and sparked after 2 seconds had elapsed. The flame produced, if any, was observed and the products of combustion analyzed. The gas mixtures were ignited by passing through them a spark from an induction coil driven by three small dry cells.

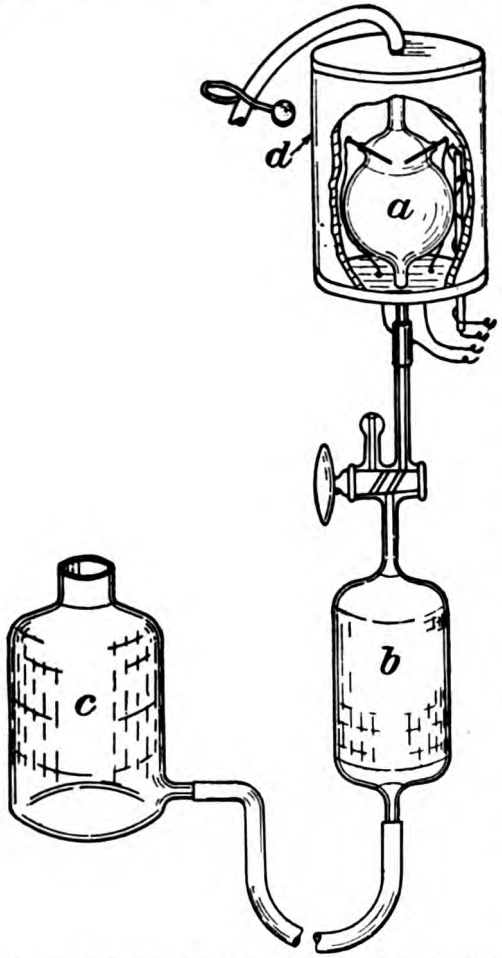

FIGURE 1.—Apparatus for determining effect of temperature and pressure on explosibility of mixtures of methane and air.

Preliminary experiments were performed with a thermocouple inserted in the explosion vessel *a* to determine the time required for the gas to attain the temperature of the oven. It was found that this time was less than 2 seconds after the introduction of the gas.

Methane was prepared from the natural gas used at Pittsburgh by fractional distillation in a vacuum at low temperatures. The natural gas was liquefied at the temperature of liquid air and as much gas pumped from it with a mercury pump as possible. The distillate was reliquefied and pumped again. The yield of pure methane was about 85 per cent of the original volume of the natural gas used.[a]

[a] For method of determining methane, see Burrell, G. A., Seibert, F. M., and Robertson, I. W., The analyses of natural gas and of illuminating gas by fractional distillations at low temperature and pressures: Tech. Paper 104, Bureau of Mines, 1915, p. 9.

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EFFECT OF INCREASING INITIAL TEMPERATURE.

The results of experiment to determine the effect of increasing the initial temperatures of the mixtures are shown in Table 1.

TABLE 1.—*Effect of increasing the initial temperature on the low limit of complete propagation of flame in methane-air mixtures.*

Test No.	Temperature.	Pressures.	Analysis of mixtures before sparking.		Analysis of mixtures after sparking.		Result as observed with the eye.	Values between which low limit lies.
			CH_4.	Air.	CH_4.	CO_2.		
	°C.	*Atmospheres.*	*Per ct.*	*Per ct.*	*Per ct.*	*Per ct.*		*Per ct.*
1....	500	1	4.47	95.53	0.58	4.08	Complete propagation.....	4.00 and 3.75
2....	500	1	4.27	95.73	.64	3.85	.....do.....	
3....	500	1	4.00	96.00	.89	3.14	.....do.....	
4....	500	1	3.75	96.25	3.45	.42	No propagation.....	
5....	500	1	3.50	96.50	3.42	.18	.....do.....	
6....	400	1	4.75	95.25	.75	3.74	Complete propagation.....	4.55 and 4.47
7....	400	.....	4.55	95.45	.65	3.93	.....do.....	
8....	400	1	4.47	95.53	4.26	.41	No propagation.....	
9....	400	1	4.25	95.73	4.12	.23	.....do.....	
10....	400	1	4.00	96.00	3.88	.20	.....do.....	
11....	400	1	3.75	96.25	3.65	.20	.....do.....	
12....	300	1	5.15	94.85	.82	4.36	Complete propagation.....	4.88 and 4.75
13....	300	1	4.88	95.02	.71	4.13	.....do.....	
14....	300	1	4.75	95.25	4.64	.13	No propagation.....	
15....	300	1	4.27	95.73	4.17	.21	.....do.....	
16....	200	1	5.15	94.85	.20	4.98	Complete propagation.....	5.15 and 4.98
17....	200	1	4.98	95.02	4.84	.18	No propagation.....	
18....	25	1	5.40	94.60	5.40	.00	.....do.....	5.46 and 5.56
19....	25	1	5.46	94.54	5.47	.00	.....do.....	
20....	25	1	5.56	94.44	.12	5.46	Complete propagation.....	

DISCUSSION OF RESULTS.

Experiments 1, 2, 3, 4, and 5 show the lower limit of complete propagation with the initial temperature at 500° C. and the initial pressure at 1 atmosphere. As far as the eye could judge, the explosion vessel was completely filled with flame when mixtures containing 4 and 4.27 per cent methane were sparked. However, a measurable amount of methane remained unburned, probably due principally to the fact that a small amount of gas was contained in the tube between the stopcock and the electric oven and did not burn. When the methane in the mixture was 3.75 and 3.50 per cent, no flame was observed, although some methane burned, as was shown by the carbon dioxide found upon analysis.

According to these results, the low limit of complete propagation is between 3.75 and 4 per cent when the initial temperature is 500° C. and the initial pressure is 1 atmosphere. At 400° C. and at atmospheric pressure a mixture containing 4.55 per cent of methane completely propagated flame, as far as could be judged by the eye,

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although 0.75 per cent methane was not burned. When the methane content was lowered to 4.47 per cent no propagation was observed. The same was true of mixtures containing 4.27, 4, and 3.75 per cent methane.

Experiments 18, 19, and 20 were performed to obtain the low limit of complete propagation of methane-air mixtures under ordinary

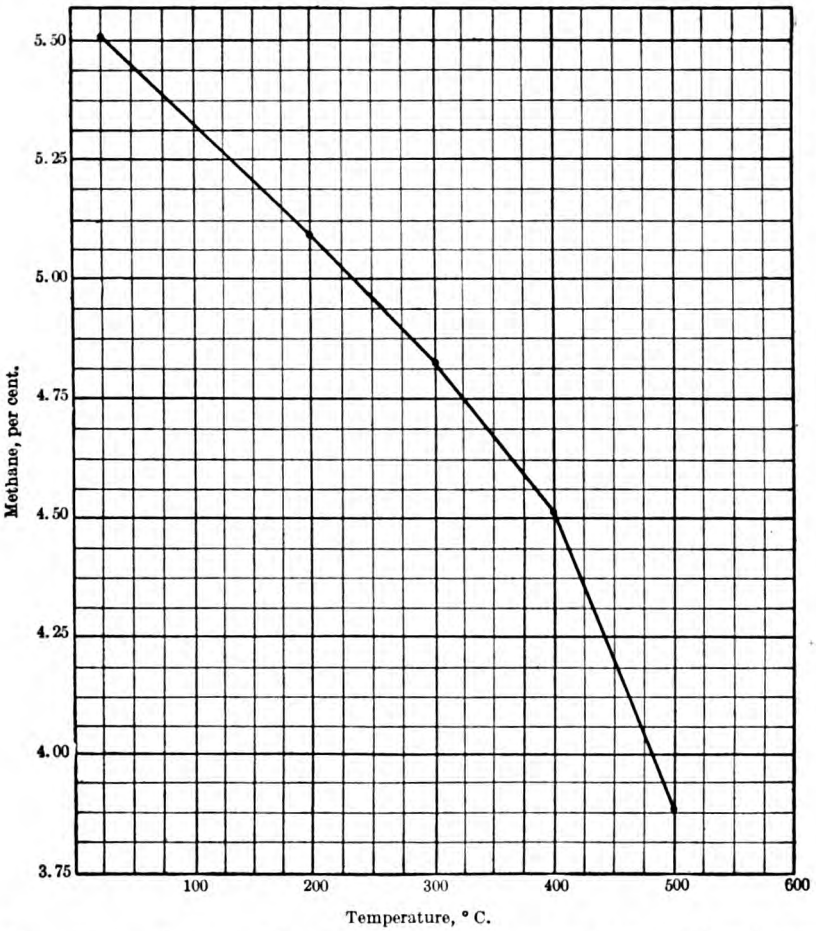

FIGURE 2.—Curve showing effect of increasing the initial temperature on the low limit of complete propagation of flame in methane-air mixtures.

conditions of temperature and of pressure. This value lies, it will be noted, between 5.46 and 5.56 per cent methane. Undoubtedly a small quantity of carbon dioxide was formed in experiments 17 and 18, but not enough to detect by the method of analysis used. In each test an analysis was made of the mixture before and after sparking. The sum of the percentages of carbon dioxide and methane

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should equal the percentage of methane present before sparking. The results of the analyses agreed quite well.

Figure 2 shows a curve plotted from the results of the experiments. The value for each temperature is the mean of two results, the percentage of methane that did not inflame, and the percentage that did.

The variation of the limits of explosibility with temperature and pressure may be explained on purely thermal grounds.

If an explosive mixture of methane and air be heated the number of collisions between molecules increases with rising temperature and the speed of reaction increases until a violent reaction and appearance of flame follow. The temperature at which this kind of a reaction takes place is called the ignition temperature. The ignition temperature of methane-air mixtures was found by Dixon and Coward [a] to be between 650° and 750° C. Slow combustion is possible, however, at temperatures below the ignition temperature, depending on the nature of the source of ignition and length of time the gas is heated. But in order that flame may be propagated throughout the gas mixture the heat of reaction of a layer of gas near the igniter must be sufficient and its rate rapid enough to raise the temperature of the adjacent layer to the ignition temperature. Obviously the higher the initial temperature the less heat will be required to raise the temperature of a given mass of the gas to the ignition temperature, and consequently the smaller is the heat of combustion and the percentage of methane that is required to furnish this quantity of heat.

EFFECTS OF EXPOSURE TO HIGH TEMPERATURES WITHOUT SPARKING.

A few experiments were performed to show the extent to which combustion would take place in various mixtures of methane and air when subjected to high temperatures without sparking. The results were as follows:

TABLE 2.—*Results of exposing methane-air mixtures to high temperatures without sparking.*

Temperature.	Pressure.	Analysis before ignition.		Analysis after ignition.		Time exposed.
		CH_4.	Air.	CH_4.	CO_2.	
° C.	*Atmospheres.*	*Per cent.*	*Per cent.*	*Per cent.*	*Per cent.*	*Minutes.*
500	1	4.27	95.73	4.21	0.10	0.5
500	1	4.27	95.73	3.89	.40	30
500	1	4.60	95.40	3.74	.90	30

The results show that no appreciable combustion occurred in the experiments shown in Table 1 between the time the gas mixtures were introduced into the pipette and the time they were sparked.

[a] Dixon, H. B., and Coward, H. F., The ignition temperature of gases: Chem. News, Mar. 19, 1909, vol. 9, p. 139.

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In the experiments described in Table 1 not longer than 2 seconds was required for the mixtures to attain the temperature of the oven after introduction into the exhausted pipette. Table 1 shows that as much as one-half minute exposure to a temperature of 500° C. resulted in only a small amount of carbon dioxide.

EFFECTS OF INCREASING THE INITIAL PRESSURE.

Some experiments were made with the apparatus shown in figure 1 to determine the effect of initial pressures higher than ordinary pressure on the explosibility of methane-air mixtures. The first experiments were made with pressures up to 5 atmospheres, obtained

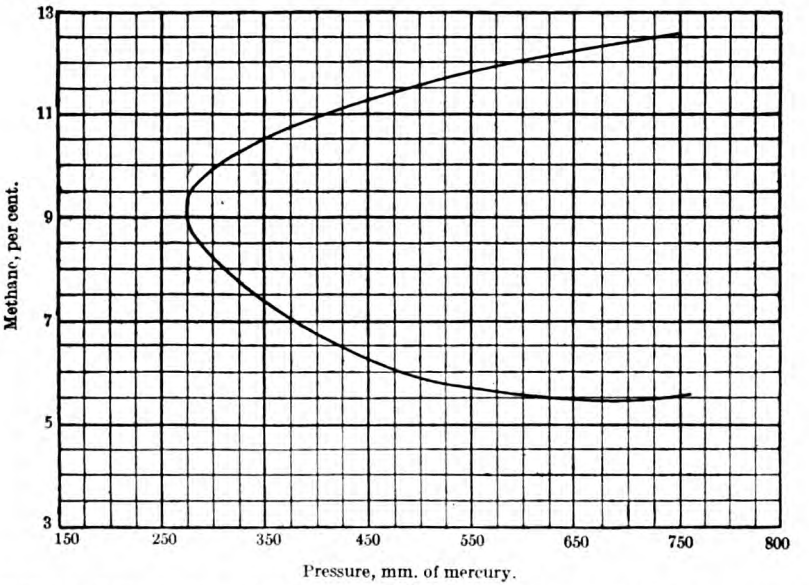

FIGURE 3.—Curve showing explosibility of methane-air mixtures at pressures less than atmospheric.

by raising the leveling bottle of the apparatus high enough to give the required pressure. It was found that increasing the initial pressure up to 5 atmospheres had no effect in changing the low limit of complete propagation. In other words, the low-limit value of about 5.5 per cent methane is true at 5 atmospheres pressure. Mr. Taffanel, director of the French experimental station at Liévin, informed the authors that in the case of hydrogen-air mixtures an initial pressure of 40 atmospheres had no effect in changing the limit of inflammability.

EFFECTS OF DECREASING THE INITIAL PRESSURE.

Experiments were performed to show the sensitiveness to ignition of various methane-air mixtures by determining the least pressures

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necessary to ignite them with a given sparking arrangement. Pressures lower than atmospheric were obtained by lowering the mercury in the leveling bottle the required distance below that in the explosion pipette.

Table 3 shows the results obtained. The second column gives the pressures. The third gives the percentage of methane, the fourth gives the percentage of air, and the fifth gives the result as observed with the eye. Enough experiments were made to draw a curve (fig. 3) representing the results.

TABLE 3.—*Results of experiments to determine explosibility of methane-air mixtures at pressures less than atmospheric.*

Test No.	Pressure, mm. of mercury.	Methane.	Air.	Result of observation.
		Per cent.	*Per cent.*	
1	550	5.59	94.41	No propagation.
2	600	5.59	94.41	Do.
3	500	5.83	94.17	Propagation.
4	450	5.83	94.17	No propagation.
5	450	6.04	93.96	Do.
6	500	6.04	93.96	Propagation.
7	450	6.13	93.87	No propagation.
8	500	6.13	93.87	Propagation.
9	370	7.03	92.97	No propagation.
10	400	7.03	92.97	Propagation.
11	320	7.87	92.13	No propagation.
12	350	7.87	92.13	Propagation.
13	300	8.50	91.50	Do.
14	250	8.50	91.50	No propagation.
15	250	9.00	91.00	Do.
16	300	9.00	91.00	Propagation.
17	300	9.50	90.50	Do.
18	250	9.50	90.50	No propagation.
19	250	10.00	90.00	Do.
20	300	10.00	90.00	Propagation.
21	350	10.30	89.70	No propagation.
22	400	10.30	89.70	Propagation.
23	350	10.75	89.25	No propagation.
24	400	10.75	89.25	Propagation.
25	450	11.00	89.00	No propagation.
26	500	11.00	89.00	Propagation.
27	450	11.50	88.50	No propagation.
28	500	11.50	88.50	Propagation.
29	500	12.00	88.00	No propagation.
30	550	12.00	88.00	Propagation.

COMMENT ON RESULTS.

It will be observed that the inflammability of the various mixtures decreased as the pressure was lowered, until it was found impossible to propagate flame in any mixture at pressures between 250 and 300 mm. of mercury.

In test 1 a mixture of methane and air containing 5.59 per cent methane did not propagate flame when the pressure was 500 mm. of mercury, but propagated flame when the pressure was 600 mm.

In test 2 a mixture containing 5.83 per cent of methane propagated flame when the pressure was 500 mm., but did not at 450 mm. The same procedure was followed in all of the tests. A certain mixture was prepared and the pressure varied to find at what pressures the mixture did and did not propagate flame. The lowest pressures at

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which propagation was recorded were 300 mm. for mixtures containing 8.5, 9, 9.5, and 10 per cent methane. In other words, the mixtures most sensitive to inflammation at low pressures lie between 8.5 and 10 per cent methane under the particular conditions of experiment adopted.

EFFECT OF VARYING SOURCE OF IGNITION.

The method of igniting the mixtures was found to have a vital influence on the results and accounts for the somewhat irregular nature of the curve. A spark from an induction coil driven by four dry cells was the source of ignition, as in the other tests recorded herein. The platinum points across which the spark jumped were one-sixteenth inch apart.

In the tests at low pressures some of the mixtures would propagate flame by the prolonged passage of the spark when an instantaneous spark would not inflame them. Instantaneous sparking was the method adopted. Some experiments were also performed in which ignition was caused by pulling apart two No. 12 gauge copper wires through which a current of 4 amperes at a pressure of 220 volts was passing. With this form of ignition a mixture of methane and air containing 9 per cent methane and under a pressure of 200 mm. propagated flame. No such inconsistencies were observed in explosion experiments at atmospheric pressure. The low limit of complete propagation was found to be practically the same with the spark from the induction coil as with a much larger electric flash.

These results show that the energy of the spark or flash is a factor of much importance, hence the curve in figure 3 represents only the least pressures at which the gas mixtures were ignited with the chosen igniting arrangement. But when the igniting arrangement is constant, or nearly so, the important factor that determines ignition is the heat of combustion of the gases. When the pressure of the gas mixture is reduced the ratio of the heat absorbed by the surroundings to the heat of combustion of the gases increases until a cooling effect is set up that is pronounced enough to prevent propagation of flame throughout the mixture.

CONCLUSIONS AS TO RESULTS OF TESTS.

When the initial temperature of methane-air mixtures is 500° C., the low limit of complete propagation of flame of the mixtures is between 3.75 and 4 per cent methane. As the initial temperature decreases from 500° C. the low limit increases until at ordinary temperatures it is about 5.5 per cent methane. Differences in the initial temperature of as much as 200° C. higher than ordinary temperature reduce the low limit from 5.50 per cent to between 4.98 and 5.15 per cent methane.

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Initial pressures as high as 5 atmospheres have no effect in changing the low limit of complete propagation of flame in methane-air mixtures. At pressures between 250 and 300 mm. of mercury mixtures of methane and air were not ignited by the igniting arrangement chosen.

The mixtures that will propagate flame at the lowest pressure contain from 8.5 to 10 per cent methane, and therefore are more sensitive to ignition than mixtures near the upper and lower limits.

The results are important in that they show that pressure and temperature conditions may vary over rather a wide range without affecting the explosibility of methane-air mixtures. Inconsistent results that have been obtained in the laboratory by different investigators on the limits of inflammation of methane-air mixtures can not be explained on the basis of slight variations in temperatures and pressures. They depend rather on the nature of the source of ignition, method of ignition, size and shape of the containing vessel, and in some cases inaccuracies in mixing and analyzing the gases.

As the low limit of complete inflammation for methane-air mixtures is not changed at pressures as great as 5 atmospheres, it can be stated that even in the deepest coal mines the low limit is not different from the limit at ordinary pressures.

PUBLICATIONS ON GAS ANALYSIS AND METHODS OF COAL MINING.

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BULLETIN 17. A primer on explosives for coal miners, by C. E. Munroe and Clarence Hall. 61 pp., 10 pls., 12 figs. Reprint of United States Geological Survey Bulletin 423.

BULLETIN 42. The sampling and examination of mine gases and natural gas, by G. A. Burrell and F. M. Seibert. 1913. 116 pp., 2 pls., 23 figs.

BULLETIN 45. Sand available for filling mine workings in the Northern Anthracite Coal Basin of Pennsylvania, by N. H. Darton. 1913. 33 pp., 8 pls., 5 figs.

BULLETIN 46. An investigation of explosion-proof mine motors, by H. H. Clark. 1912. 44 pp., 6 pls., 14 figs.

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BULLETIN 56. First series of coal-dust explosion tests in the experimental mine, by G. S. Rice, L. M. Jones, J. K. Clement, and W. L. Egy. 1913. 115 pp., 12 pls., 28 figs.

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TECHNICAL PAPER 75. Permissible electric lamps for miners, by H. H. Clark. 1914. 21 pp., 3 figs.

TECHNICAL PAPER 76. Notes on the sampling and analysis of coal, by A. C. Fieldner. 1914. 59 pp., 6 figs.

TECHNICAL PAPER 77. Report of the Committee on Resuscitation from Mine Gases, by W. B. Cannon, George W. Crile, Joseph Erlanger, Yandell Henderson, and S. T. Meltzer. 1914. 36 pp., 4 figs.

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